Section 2 Optics and Devices

Chapter 1 Optics and Quantum Electronics.

Chapter 2 Infrared Nonlinear Optics.

Chapter 1. Optics and Quantum Electronics

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1.1 The Nonlinear Waveguide Interferometer

Sponsors

National Science Foundation
(Grant EET 87-00474)
Joint Services Electronics Program
(Contracts DAAL03-86-K-0002
and DAAL03-89-C-0001)

Project Staff

Professor Hermann A. Haus, Professor James G. Fujimoto, Keren Bergman, Michael J. LaGasse, Dilys L. Wong

The principal difficulty with fiber interferometers is their susceptibility to thermal and acoustic fluctuation effects. A novel design of a nonlinear waveguide interferometer that avoids these effects was proposed by Matasaki Shirasaki from Fujitsu, while he was a visiting scientist within RLE[2] It is shown in figure 1. The spatial separation of two interfering pulses in a two-arm interferometer is replaced by temporal separation in one single arm. The interferometer is insensitive to acoustic and thermal fluctuations on a time scale which is long in comparison to that of the pulse separation.

We have reported pulse switching with this interferometer.[3] The extraordinary stability of the interferometer was confirmed experimentally. The interferometer is so well balanced that it does not need a stabilization circuit. Work on the use of this interferometer and other generic forms of the system as all-optical switches is continuing. Variations of the design promise to provide "squeezed" optical pulses for use of increased sensitivity in interferometric measurements and in fiber gyros (see section 1.4, " Squeezing of Pulses" on page 64). A modification of the interferometric scheme has proven useful for the determination of

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[1] Polaroid Corporation.

[2] H.A. Haus, M. Shirasaki, and D.L. Wong, "A Nonlinear Fiber Interferometer and Logic Gate." Paper presented at CLEO '87, Baltimore, Maryland, April 27, 1987.

[3] M.J. LaGasse, D.L. Wong, J.G. Fujimoto, and H.A. Haus, "Femtosecond Pump-Probe Interferometry." Paper presented at IQEC '88, Tokyo, Japan, July 1988; M.J. LaGasse, D.L. Wong, J.G. Fujimoto, and H.A. Haus, "Ultrafast Switching with a Single Fiber Interferometer," *Opt. Lett.* 14 (6):311 (1989).

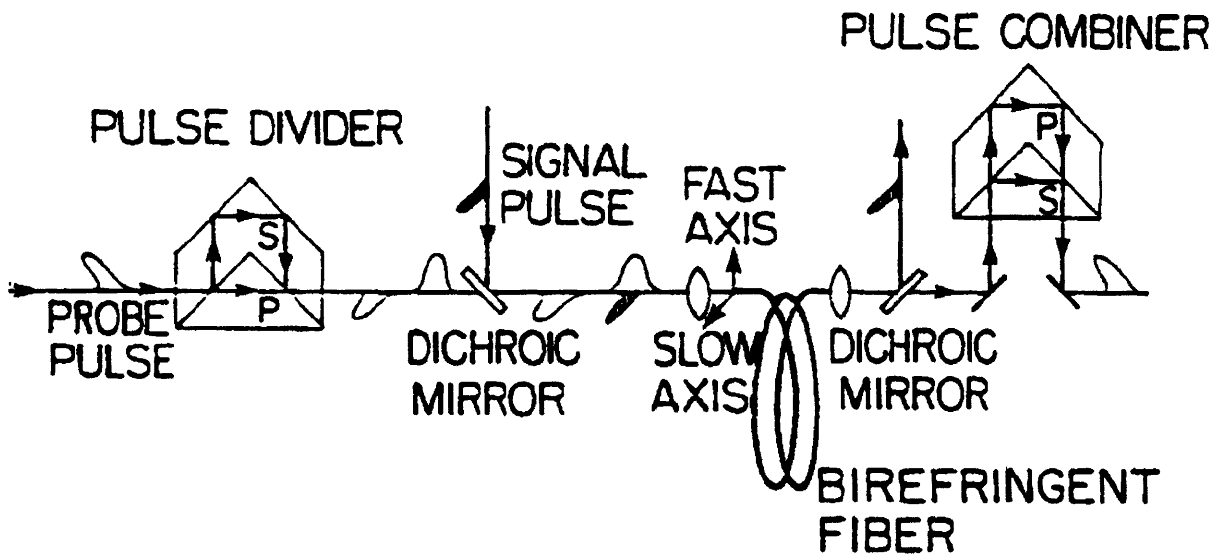

Figure 1. Schematic of single-fiber nonlinear interferometer.

high-speed optical waveguide responses (in section 1.5, " Multiple Quantum Well Semiconductor Waveguide Optical Devices" on page 64).

1.2 Picosecond Optical Signal Sampling

Sponsor

National Science Foundation
(Grant EET 87-00474)

Project Staff

Kristin K. Anderson, Professor Hermann A. Haus, Lynne A. Molter, Weiping Huang

Further progress has been made on the theory of mode-coupling and methods for the evaluation of waveguide dispersion characteristics. A self-consistent theory of coupling of nonorthogonal modes in tapered structures has been developed[4] that corrected erroneous approaches in the literature.[5]

Large extinction ratios and cross-talk suppression are important attributes of optical switches. The conventional coupled mode theory does not give correct results when the coupling is of the magnitude commonly used in coupler design. We have investigated methods for improved crosstalk suppression in two-guide couplers and $\delta\beta$ couplers taking the effects of strong coupling into account.[6]

[4] H.A. Haus and W.P. Huang, "Mode Coupling in Tapered Structures," *J. Lightwave Tech.*, to be published.

[5] A. Hardy, M. Osinski, and W. Streifer, "Application of Coupled-Mode Theory to Nearly Parallel Waveguide Systems," *Electron. Lett.* 22:1249-1250 (1986); M.A. McHenry and D.C. Chang, "Coupled-Mode Theory of Two Nonparallel Dielectric Waveguides," *IEEE Trans. Microwave Theory Tech.* 32:1469-1475 (1984); C. Vassallo, "About Coupled-Mode Theories for Dielectric Waveguides," *J. Lightwave Tech.* 6:294-303 (1988).

[6] J.P. Donnelly, L.A. Molter, G.S. Hopcraft, R.E. Smith, and H A. Haus, "Extinction Ratio in Optical Two-Guide $\delta\beta$ Couplers," presented at OFC, Houston, Texas, February 1989; M.J. Robertson, S. Richie, and O.P. Dayan, "Semiconductor Waveguides: Analysis of Optical Propagation in Single Rib Structures and Directional Couplers," *IEEE Proc.* 132:336-342 (1985).

Power transfer in grating coupled optical waveguides has been studied and some peculiarities of the output chracteristic identified and explained.[7] A program for the evaluation of dispersion characteristics of ridge waveguides has been implemented. It can be run on a PC and is fast. The results are in good agreement with other current programs that are very computation intensive.[8]

1.3 Solitons

Sponsors

Charles Stark Draper Laboratory
(Grant DL-H-285408)
Joint Services Electronics Program
(Contracts DAAL03-86-K-0002
and DAAL03-89-C-0001)

Project Staff

Professor Hermann A. Haus, Professor Erich P. Ippen, Janice M. Huxley, Yincheih Lai, Ling-Yi Liu, John D. Moores

A soliton laser has been operating for some time in our laboratory. Along with other researchers, we found that the system operates in the positive dispersion regime of the fiber. Soliton operation is a special case of a more general operating principle that we have dubbed Additive Pulse Mode-Locking (APM). Additional details are given in the section on APM. In this section, we are reporting on work that is specifically aimed at studying various aspects of solitons in fibers.

The interaction of two orthogonally polarized "soliton" pulses in a birefringent fiber has been studied numerically. Such pulses do not behave as conventional fiber solitons in a collision. The numerical studies revealed under what conditions perturbation approximations hold, i.e., the pulses do behave like interacting solitons.[9] This finding is relevant to all-optical switching with orthogonally polarized pulses.

Experimental work began last year on the formation of solitons in fibers pumped by pulses that provide Raman gain. In anticipation of the operation of a ring fiber Raman soliton laser, that is known to be prone to produce pulses with a background (that appears as a pedestal on the autocorrelation trace), we have invented an interferometric system that removes the pedestal when the radiation is passed through it.[10] In the coming year, we shall construct and test the system.

When solitons are used for measurements at the quantum limit, it is important to have a reliable quantum theory of solitons. We have developed two approaches, one based on the Poisson bracket formulation of the equations of motion of the soliton parameters,[11] the other on a direct integration of the equations of motion in the Schroedinger formulation of the problem.[12]

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7 W.P. Huang and H.A. Haus, "Power Exchange in Grating-Assisted Couplers," *J. Lighwave Tech.*, to be published.

8 J.P. Donnelly, H.A. Haus, and L.A. Molter, "Crosspower and Crosstalk in Waveguide Couplers," *J. Lightwave Tech.* 6:257 (1988).

9 J.D. Moores, *Collisions of Orthogonally Polarized Solitary Waves.* S.M. thesis, Dept. of Electr. Eng. and Comp. Sci., MIT, 1989.

10 Invention: patent application is in progress.

11 H.A. Haus, K. Watanabe, and Y. Yamamoto, "Quantum Nondemolition Measurement of Optical Solitons," *J. Opt. Soc. Am. B,* submitted for publication.

12 Y. Lai and H.A. Haus, "Quantum Theory of Solitons in Optical Fibers. I. Time Dependent Hartree Approximation," *Phys. Rev. A,* submitted for publication; Y. Lai and H.A. Haus, "Quantum Theory of Solitons in Optical Fibers. II. Exact Solution," *Phys. Rev. A,* submitted for publication.

1.4 Squeezing of Pulses

Sponsors

Charles Stark Draper Laboratory
(Grant DL-H-2854018)
National Science Foundation
(Grant EET 87-00474)

Project Staff

Keren Bergman, Yincheih Lai, Matasaka Shirasaki, Professor Hermann A. Haus

Squeezing of quasi-cw radiation has now been achieved in many laboratories. Squeezing of optical pulses of large bandwidth should avoid some of the classical sources of noise that plague quasi-cw squeezing. Our scheme for squeezing of pulses[13] has advantages over other systems because it 1) is broadband, 2) separates the pump from the squeezed radiation, and 3) utilizes a version of the single fiber interferometer described in section 1.1, " The Nonlinear Waveguide Interferometer" on page 61, while taking advantage of its stability.

The system is shown in figure 2. The pump pulse is split into two countertraveling versions of itself by the beam splitter and injected into the ring interferometer. If the interferometer were linear, the pump pulse would be fed out through the "input port," and zero-point fluctuations would emerge at port (2). The nonlinearity of the interferometer causes photons to be deflected into port (2) coherently with the zero-point fluctations injected at port (2). This effect results in squeezed, pulsed, zero-point fluctuations emerging at port (2). The balanced detector is used to detect the squeezed pulses, using a portion of the returning pump pulse as the local oscillator.

One remarkable feature of the system is that it can be converted into a fiber gyro with only one modification: a nonreciprocal phase shifter can be introduced into the ring. The amount of phase shift used to balance the

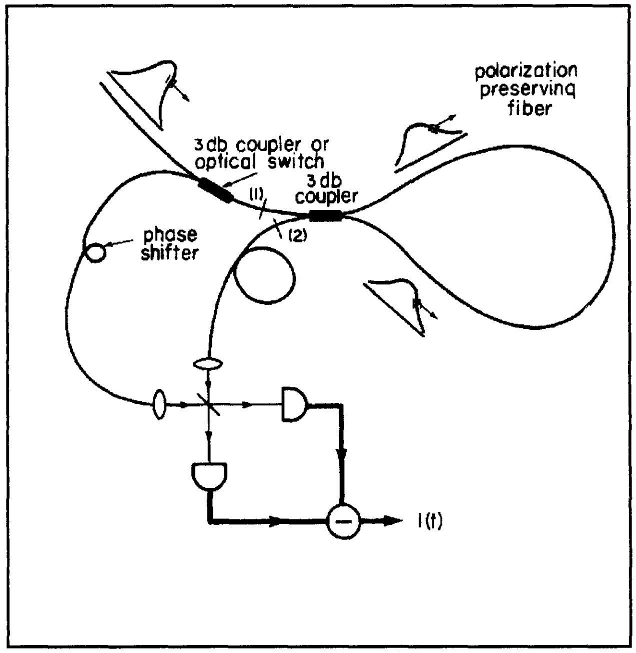

Figure 2. Schematic of experimental setup.

Sagnac effect is the gyro signal. We intend to pursue this basic idea in the future.

1.5 Multiple Quantum Well Semiconductor Waveguide Optical Devices

Sponsors

Joint Services Electronics Program
(Contracts DAAL03-86-K-0002
and DAAL03-89-C-0001)
National Science Foundation
(Grant EET 87-03404)

Project Staff

Kristin K. Anderson, Stuart D. Brorson, Mary Phillips, Professor Hermann A. Haus, Professor Clifton G. Fonstad, Jr.

Multiple quantum well (MQW) semiconductor modulators and switches have important applications in optical signal processing systems. The thrust of our work is in the fabrication and high-speed characterization of MQW waveguide structures.

[13] M. Shirasaki and H.A. Haus, "Broadband Squeezing with Cotraveling Waves," *Topical Meeting on Nonlinear Guided Wave Phenomena: Physics and Applications,* Houston, Texas, February 2-4, 1989.

A new method of waveguide fabrication has been developed and tested in GaAs/AlGaAs MQW layers. It uses disordering of the quantum well layers by ion bombardment to produce low index regions adjacent to the higher index quantum well regions. It was found that nitrogen ions are best suited for the purpose because they produce disordered regions with relatively low optical loss.[14] Such waveguides are particularly well suited for interferometric switching with the optical Stark effect.

In addition to the well known GaAs/AlGaAs system, we are investigating MQW structures in InGaAs-InAlAs. Very good structures have been grown as verified by means of transmission spectroscopy. Tests of the subpicosecond response of the structures the soliton laser as a source have been initiated and are being continued.

1.6 Additive Pulse Mode Locking

Sponsors

Joint Services Electronics Program
(Contracts DAAL03-86-K-0002 and DAAL03-89-C-0001)
National Science Foundation
(Grant ECS 84-06290)
U.S. Air Force - Office of Scientific Research
(Contract F49620-88-C-0089)

Project Staff

Katherine L. Hall, Ling-Yi Liu, Jannik Mark, Professor Hermann A. Haus, Professor Erich P. Ippen

Work in our laboratory with a synchronously-pumped color-center laser (KCl:Tl at 1.5 μm), coupled to a fiber resonator has resulted in the identification of a new mode-locking mechanism: additive pulse mode locking. In previous experiments at AT&T Bell Laboratories,[15] it was shown that ultrashort pulses could be generated with such a system operating under conditions where the pulses were shaped by soliton propagation effects in the fiber. Our work shows that soliton shaping effects are not necessary for femtosecond pulse generation. Pulse shortening is produced by the amplitude interference effects at the coupling mirror between the laser and fiber resonators. This additive pulse mode locking (APM) is as least as effective as, if not more effective, than when the fiber dispersion is positive than when it is negative.

The principal of APM was demonstrated experimentally[16] by measuring output pulse durations as the relative optical phase of the two pulses as the coupling mirror was varied. Nonlinearity in the fiber always results in a retardation of the optical phase of the pulse near maximum intensity. When this phase modulated pulse is coherently added to the one in the laser resonator, pulse shortening can be produced. Our experiments showed that the phase in the wings of the pulse from the fiber must lead the other in phase, so that improved addition occurs at the peak. With a laser that produced 20 ps pulses without coupling to the fiber cavity, pulses as short as 100 fs were generated with optimum adjustment of the coupling phase. Similar results were achieved with both positive and negative dispersion in the fiber.

In addition to the experimental demonstration of APM, we have formulated, and submitted for publication, a theoretical treatment of mode locking under these conditions. Work continues toward a more fully developed theoretical analysis, the generation of shorter pulses, and extention to other broadband laser systems.

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[14] K.K. Anderson, J.P. Donnelly, C.A. Wang, J.D. Woodhouse, and H.A. Haus, "Compositional Disordering of GaAs/AlGaAs Multiple Quantum Wells Using Ion Bombardment at Elevated Temperatures," *Appl. Phys. Lett.* 53:1632 (1988).

[15] L.F. Mollenauer and R.H. Stolen, *Opt. Lett.* 9:13 (1984).

[16] J. Mark, L.Y. Liu, K.L. Hall, H.A. Haus and E.P. Ippen, *Opt. Lett.* 14:48 (1989).

1.7 Femtosecond Pulse Propagation and Amplification in GaAs Laser Diodes

Sponsors

Joint Services Electronics Program
(Contracts DAAL03-86-K-0002 and DAAL03-89-C-0001)
National Science Foundation
(Grant ECS 84-06290)
U.S. Air Force - Office of Scientific Research
(Contract F49620-88-C-0089)

Project Staff

Stuart D. Brorson, Charles T. Hultgren, Morris P. Kesler, Professor Erich P. Ippen

We have studied both the spectral dynamics of gain saturation and group velocity dispersion in GaAs laser diodes with femtosecond pulses from a tunable dye laser system. Previous experiments[17] showed that nonequilibrium carrier heating dominates the gain compression on a sub-picosecond timescale and suggested that spectral hole burning relaxes in less than 50 fs. Those experiments were performed with femtosecond pump and probe pulses at the same wavelength. Recently, we have extended our experiments to allow probing at wavelengths different from that of the pump by using up-conversion gating of cw probing beams from other diode lasers. Results obtained in this manner[18] reveal simultaneous, femtosecond gain compression at wavelengths throughout the gain bandwidth, comfirming our previous prediction.

Group velocity dispersion in semiconductor lasers and amplifiers limits the resolution of femtosecond measurement, influences nonlinear wave mixing, and affects ultrashort pulse generation by mode locking. Estimates of these effects have to-date relied upon studies of variations in Fabry-Perot laser mode spacings. With femtosecond pulses we are now able to measure group round-trip delay in GaAs diodes directly in the time domain by cross-correlation of the output with the input. Multiple reflections within a diode result in a transmitted train of pulse echoes that can be used to improve the accuracy of the measurement. By observing the change in round-trip delay with wavelength we can determine the dispersion. Results indicate a change in group index with wavelength of $-2.9\ \mu m^{-1}$ and critical pulsewidth (that which doubles upon propagation through the amplifier) of 75.[19]

1.8 Ultrafast Dynamics in InGaAsP Devices

Sponsors

Joint Services Electronics Program
(Contracts DAAL03-86-K-0002 and DAAL03-89-C-0001)
U.S. Air Force - Office of Scientific Research
(Contract F49620-88-C-0089)

Project Staff

Katherine L. Hall, Jannik Mark, Ling-Yi Liu, Professor Erich P. Ippen

Semiconductor optical amplifiers and other active waveguide devices are receiving increasing attention for potential application to broadband communication and signal processing systems. Of particular interest are InGaAsP components that operate in the fiber communication bands of 1.3 μm and 1.5 μm. Much is known about their linear and small signal characteristics, much less about their dynamics and nonlinear behavior. This year, with femtosecond pulses from our APM color-center laser (tunable from 1.48 to 1.54 μm), we have been able to begin the first studies of ultrafast dynamics in InGaAsP laser amplifiers. Early results indicate significant differences in behavior between InGaAsP and AlGaAs.

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[17] M.P. Kesler and E.P. Ippen, *Appl. Phys. Lett.* 51:1765 (1987).

[18] M.P. Kesler and E.P. Ippen, *Electron. Lett.* 24:1102 (1988).

[19] M.P. Kesler and E.P. Ippen, to be published.

Our experiments utilize pump and probe pulses of the small wavelength but of orthogonal polarization. By varying either the pump-probe wavelength or the current to the laser amplifier, we are able to study dynamic, induced charges in transmission under conditions of gain, loss or nonlinear transparency. It is therefore possible to separate dynamic effects associated with population changes (such as hole burning) from those that are not (such as carrier heating). In AlGaAs devices only the latter are observed. In our quaternary amplifiers, we observe carrier heating with a recovery time of 650 fs. In addition, however, there are strong components that appear to relate to two-photon absorption (instantaneous recovery) and nonequilibrium changes in carrier distributions ($\tau \cong 250$ fs). Work continues toward understanding the physical mechanisms behind these nonlinearities and toward evaluating their potential use for optical switching.

1.9 Femtosecond Laser Systems

Sponsors

AT&T Bell Foundation
National Science Foundation
(Grant ECS 85-52701)
Joint Services Electronics Program
(Contracts DAAL03-86-K-0002 and DAAL03-89-C-0001)
U.S. Air Force - Office of Scientific Research
(Contract F49620-88-C-0089)

Project Staff

Professor James G. Fujimoto, Beat Zysset, Jhypyng Wang, Michael J. LaGasse, Robert W. Schoenlein, and Morrison Ulman, in collaboration with Dr. Peter Schulz

1.9.1 Introduction

The development of new femtosecond laser generation and measurement technology is of central importance for studies of ultrafast phenomena. The emphasis of our program in femtosecond laser technology is twofold. First, the laser systems form the basis for research on high speed processes in materials, devices, and systems. In principle, the temporal resolution which may be achieved in ultrafast studies is limited only by the shortest laser pulse which can be generated. In addition, the development of new laser technology is essential in itself for widespread applications of high speed optics to communications and signal processing.

1.9.2 Multistage High Repetition Rate Femtosecond Amplifiers

The optimum laser system for ultrafast studies would produce high intensity, ultrashort pulses over a broad range of wavelengths and at a high repetition rate. The development of such a system requires both a laser source for short pulses as well as amplifiers for increasing pulse energy.

One of our present laser systems is based on a colliding pulse mode-locked ring dye laser as a source and a copper vapor laser pumped amplifier.[20] The laser operates at 620 nm and produces pulse durations as short as 60 fs. The repetition rate of our system (8 kHz) is determined by the copper vapor laser pump. This high repetition rate is important for achieving high measurement sensitivity using signal averaging and lock-in detection.

Currently, we are developing a new multistage copper vapor laser pumped dye cell amplifier to increase pulse energy, decrease pulse duration, and provide tunability over a range of wavelengths. The multistage amplifier is more efficient and experimentally convenient than earlier designs. The multistage design permits mode matching between the amplified and pump beams. The design concept is analogous to that used in conven-

[20] W.H. Knox, M.C. Downer, R.L. Fork, and C.V. Shank, "Amplified Femtosecond Optical Pulses and Continuum Generation at 5 kHz Repetition Rate," *Opt. Lett.* 9:552-554 (1984).

tional microwave design. The initial stages are optimized for high gain, while the final stage is optimzed for power extraction.

The overall gain of the system is as high as 10^5. The amplifier can produce pulse energies in excess of 25 μJ. For pulses of 60 fs duration, this corresponds to peak intensities of several hundred megawatts. Since femtosecond pulses permit the generation of high peak intensities with modest pulse energies, nonlinear processes may be used to change the wavelength of the pulses or perform pulse compression.

We are currently investigating new amplifier topologies which cascade successive gain stages with nonlinearities. For example, by using self phase modulation in an optical fiber, the bandwidth of the pulses may be increased. If this is followed by a dispersive delay line or phase filter, then the pulses may be compressed. Construction of a system which has a pulse compressor between successive gain stages is yield pulses shorter than 20 fs with several hundred megawatt peak intensity. Other nonlinear optical techniques should result in the generation of wavelength tunable pulses.

1.9.3 Diode Pumped Nd:YAG Amplifier

The investigation of ultrafast dynamics in optical materials and devices requires a laser system with femtosecond pulse durations, tunable wavelengths, and high repetition rates. A wide range of ultrafast phenomena, such as electron-hole dynamics in semiconductors, require sources with pulses durations on the order of less than 50 fs. Many processes have a photon energy threshold dependence, thus, tunability is highly desirable. A high repetition rate system enables the investigator to use powerful signal averaging techniques such as locking detection.

We have recently demonstrated a high repetition rate femtosecond laser system using a laser-diode pumped *Q*-switched Nd:YAG laser.[21] The laser-diode pumped Nd:YAG is a promising and rapidly emerging technology which has the potential for producing a compact and versatile femtosecond laser technology.

Our tunable laser femtosecond laser system was used for these experiments. This system generates wavelength tunable 300 fs laser pulses using a cascade pulse compression technique similar to that developed by Kafka and Baer.[22] A mode-locked Nd:YAG laser which generates 90 ps pulses at 1.06 μm is pulse compressed to 5 ps using a single mode optical fiber and diffraction grating pair. These pulses are subsequently frequency doubled and used to synchronously pump a dye laser. By using different laser dyes, femtosecond pulses can be obtained in the wavelength range 580 to 980 nm.

Wavelength tunable, 300 fs pulses generated by our synchronously-pumped dye laser system using the dye rhodamine 6G were amplified in a dye jet pumped by a frequency-doubled, laser-diode pumped, *Q*-switched, Nd:YAG laser. The small signal gain was 200 and the saturated gain was 70 at a repetition rate of 800 Hz. Pulse energies as high as 40 nJ were achieved. These high pulse energies enabled pulse compression using an optical fiber and diffraction gratings. Pulse durations as short as 30 fs were obtained at a wavelength of 580 nm. The pulse durations remained less than 50 fs throughout the tuning range of 580 to 620 nm.

This system is a highly reliable source of tunable femtosecond pulses suitable for a wide range of experimental investigations such as hot carrier dynamics and intervalley scattering in AlGaAs. Extending this technique to the near infrared, would permit bandedge studies in AlGaAs which are relevant to ultrafast all-optical switching.

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[21] B. Zysset, M.J. LaGasse, J.G. Fujimoto, and J.D. Kafka, "High Repetition Rate Femtosecond Dye Amplifier Using a Laser-Diode Pumped Neodymium-YAG Laser," *Appl. Phys. Lett.* 54:496 (1989).

[22] J.D. Kafka and T. Baer, "A Synchronously Pumped Dye Laser Using Ultrashort Pump Pulses," *Proc. Soc. Photo-Opt. Instrum. Eng.* 533:38 (1985).

1.9.4 Variable Pulse Duration and Pulse Synthesized Laser

Although pulse lasers have found applications in many research areas, they have never been versatile pulse sources compared to pulse generators in low frequency electronics. Both the pulse duration and the pulse repetition rate are predetermined by the processes which generate the pulses, and cannot be varied without redesign and reconstructing the laser. In many research areas such as laser surgery, laser fusion, time-resolved spectroscopy and optical communication, there has been great demand for the technology of general waveform synthesis on high power laser pulses.

We are constructing a solid state laser system which features high pulse energy, variable pulse duration and repetition rate. The system consists of a high power mode-locked Nd:YLF laser, a pulse compression/stretching stage,[23] a single pulse selector, and a Nd:Phosphate glass regenerative amplifier.[24] The regenerative amplifier is a ring cavity amplifier, which serves both as the pulse amplification and storage ring.

Variable pulse duration is produced using an optical pulse compression/stretching technique. A mode-locked pulse train from the Nd:YLF laser is first sent through a long optical fiber. Frequency dispersion and self-phase modulation in the fiber produce a linear frequency chirp in the laser pulse, which can be compressed by a grating pair. By varying the distance between the grating pair, the degree of compression, the pulse duration can be adjusted. The combination of this technique with frequency and phase filtering can permit the generation of an arbitrary synthesized laser pulse shape.

Variable pulse repetition rate is achieved by employing electro-optical switching technique in the regenerative amplifier. A Glan prism in the amplifier cavity switches the pulse in and out according to the polarization, and a fast high voltage controlled Pockels cell is used to rotate the polarization. After being switched in, the optical pulse can be stored and amplified in the cavity, and switched out at a desired time.

Since the Nd:Phosphate glass laser has a very high saturation level, it can support much higher energies than other solid state lasers. Furthermore, its bandwidth is much broader than other solid state lasers and allows amplification of subpicosecond pulses.[25] This is important for laser fusion research in which both high pulse energy and high peak intensity are essential.

One of the problems in the high energy glass laser is the wavefront instability induced by self-focusing. When the intensity is high enough, the nonlinear interaction between the glass and the light induces higher index of refraction in the region of higher energy density. This variation of index of refraction on the wavefront acts as converging lenses, which amplify wavefront fluctuations. The effect becomes more pronounced as the total length of travel in the glass increases. We have been investigating the techniques to overcome this problem. One technique employed in our system is placing the pulse stretcher before and the compressor after the amplifier, so that the peak intensity in the amplifier is reduced.

One of the major applications of this laser system is in laser microsurgery. Details of the application will be presented in section 1.12, "Laser Medicine" on page 74. The

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[23] W.J. Tomlinson, R.H. Stolen, and C.V. Shank, "Compression of Optical Pulses Chirped By Self-Phase Modulation in Fibers," *J. Opt. Soc. Am. B* 1:139 (1984).

[24] L. Yan, J.-D. Lin, P.-T. Ho, C.H. Lee, and G.L. Burdge, "An Active Mode-Locked Continuous Wave Nd:Phosphate. Glass Laser Oscillator and Regenerative Amplifier," *IEEE J. Quant. Electron.* 24:418 (1988); T. Sizer and I.N. Duling, "Neodymium Lasers as a Source of Synchronized High-Power Optical Pulses," *IEEE J. Quant. Electron.* 24:404 (1988); P. Bado, M. Bouvier, and J.S. Coe, "Nd:YLF Mode-Locked Oscillator and Regenerative Amplifier," *Opt. Lett.* 12:319 (1987).

[25] W.J. Tomlinson, R.H. Stolen, and C.V. Shank, "Compression of Optical Pulses Chirped By Self-Phase Modulation in Fibers," *J. Opt. Soc. Am. B* 1:139 (1984).

essential point is: in laser microsurgery, tissue incision can be made by the plasma generated from a short laser pulse at the tissue surface.[26] By reducing the pulse energy while keeping the peak intensity above the plasma generating threshold, the size of the plasma, therefore the size of the ablation, can be reduced. However, as the pulse energy is decreased, the ablation speed is also slowed down as a trade-off.[27] For different operations the optimal pulse duration is different. Our system, with its capability of varying the pulse duration and repetition rate, allows the study of laser tissue interaction as a function of pulse duration and repetition rate. This study will provide not only the optimal operation points in laser microsurgery, but also the information for improving the current prototype of short pulse laser scalpel.

1.9.5 Short Pulse Generation with Ti:Al_2O_3 Solid State Laser Technology

Titanium sapphire (Ti:Al_2O_3) is an attractive laser material which has high energy storage, high damage threshold, and a large gain bandwidth.[28] High power, in excess of 750 mW, active mode-locked and continuous wave Ti:Al_2O_3 lasers have been demonstrated at room temperature.[29] The tuning range of the laser[29] extends from 670 nm to 1 μm. We have begun a collaborative program with Lincoln Laboratory to investigate short pulse generation in Ti:Al_2O_3.

Picosecond and femtosecond pulse generation is intrinsically very difficult with many of the new solid state laser materials. While the low gain cross section of these materials permits high energy storage, this makes mode locking or short pulse generation extremely difficult. Traditional short pulse generation techniques have proved inadequate for many of these new solid state systems. Recently, however, a new pulse shortening technique which uses a nonlinear external cavity has been demonstrated. Pulse width reduction by factors of 10 to 100 has been achieved in color center lasers.[30]

This technique thus uses external feedback to shape the laser pulse directly. The effect of the nonlinear cavity is to self-phase modulate the pulse so that the pulse's phase at the peak has been shifted relative to the phase at the pulse's edges. The chirped pulse is then returned to the main laser cavity with an appropriate phase such that main cavity pulses and external cavity pulses interfere constructively at the peak and destructively at the wings. Because this pulse shaping mechanism is not inherently dependent on wavlength, it can be used in conjunction with tunable solid state lasers. Our objective is to apply this pulse shortening technique to an actively mode-locked Ti:Al_2O_3 system.

Working in collaboration with Dr. Peter Schulz at MIT Lincoln Laboratory, we have constructed a room temperature Ti:Al_2O_3 laser capable of delivering 1.3 watts cw with pump conversion efficiency greater than 10 percent. The tuning range extends from 690 nm to 920 nm. We have actively mode-locked the laser to produce 40 ps pulses with 1 watt average power. With the use of a nonlinear external cavity, we have demonstrated pulse shortening to 2 ps.[31] We antic-

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[26] B. Zysset, J.G. Fujimoto, and T.F. Deutsch, "Time-Resolved Measurements of Picosecond Optical Breakdown," *Appl. Phys.* 48:139 (1989).

[27] D. Stern, R.W. Schoenlein, C.A. Puliafito, E.T. Dobi, R. Birngruber, and J.G. Fujimoto, to be published.

[28] P.F. Moulton, "Spectroscopic and Laser Characteristics of Ti:Al_2O_3," *J. Opt. Soc. Am. B* 3:125-133 (1986).

[29] J.D. Kafka, C.J. Culfrey, and T. Baer, "Mode-Locked Continuous Wave Titanium Sapphire Laser," *Ultrafast Phen.* PD3:65-66 (1988).

[30] P.N. Kean, X. Zhu, D.W. Crust, R.S. Grant, N. Langford, and W. Sibbertt, "Enhanced Mode Locking of Color-Center Lasers," *Opt. Lett.* 14:39-41 (1989).

[31] J. Goodberlet, J. Wang, J.G. Fujimoto, and P. Schulz, "Modelocked Ti:Al_2O_3 Laser with a Nonlinear Coupled External Cavity," paper to be presented at CLEO '89, Baltimore, Maryland, April 1989.

ipate further shortening with optimization of parameters. We also plan to assess system stability and investigate techniques of pulse compression and frequency doubling for the pulse shortened $Ti:Al_2O_3$ laser.

1.9.6 Copper Vapor Laser Pumped $Ti:Al_2O_3$ Amplifier

Working in collaboration with Dr. Peter Schulz at MIT Lincoln Laboratory, we have developed a titanium-sapphire ($Ti:Al_2O_3$) femtosecond amplifier. $Ti:Al_2O_3$ has a very wide gain bandwidth (200 nm),[32] and thus has excellent potential for amplifying femtosecond laser pulses. This system has demonstrated gain in excess of 10^7, and pulse durations of less than 300 fs at 790 nm.

The amplifier is pumped by 5 mJ pulses from a copper vapor laser and is injection-seeded by a synchronously-pumped Nd:YAG/dye laser system. This laser system employs two stages of pulse compression to generate pulses of a few hundred femtoseconds duration which can be tuned over the entire gain bandwidth of the $Ti:Al_2O_3$ amplifier. A single femtosecond pulse from the dye laser system is selected using an acousto-optical modulator, and feedback from the amplifier is suppressed using a Faraday isolator.

The amplifier consists of a 7 mm $Ti:Al_2O_3$ crystal positioned between two mirrors, with the pump beam entering through one of the mirrors (dichroic) and the injected femtosecond pulses entering through the opposite end mirror. A telescope arrangement is employed to match the seed pulses to the mode of the amplifier cavity. Injected pulses of $\sim$ 400 fs duration are amplified to energies in excess of 1 μJ ($\sim 10^7$ gain). These pulses are then compressed to 275 fs. This demonstrates the feasibility of using $Ti:Al_2O_3$ as a gain medium in high energy broadband femtosecond amplifiers.

The combination of a short pulse oscillator and amplifier in $Ti:Al_2O_3$ would result in a powerful all solid state laser system with the potential for replacing a large class of dye lasers currently used in ultrafast measurements. The broad bandwidth of this laser material as well as its high energy storage should provide pulse duration, pulse energy, and pulse shape synthesis performance superior to current dye laser systems.

1.10 Femtosecond Studies in GaAs

Sponsors

AT&T Bell Foundation
National Science Foundation
(Grant ECS 85-52701)
Joint Services Electronics Program
(Contracts DAAL03-86-K-0002
and DAAL03-89-C-0001)
U.S. Air Force - Office of Scientific Research
(Contract F49620-88-C-0089)

Project Staff

Professor James G. Fujimoto, Beat Zysset, Michael J. LaGasse, Kristen K. Anderson, Robert W. Schoenlein, Professor Hermann A. Haus

1.10.1 Carrier Dynamics and Intervalley Scattering

The investigation of hot carrier dynamics in AlGaAs is relevant to electronic and optoelectronic devices which depend on high speed transient carrier phenomena. Fundamental carrier scattering processes in AlGaAs occur on a 50 fs time scale. Thus, optical techniques are required for the characterization of these ultrafast processes. Electron scattering to the X and L satellite valleys occurs on a femtosecond time scale and gives rise to negative differential resistivity

[32] W.H. Knox, M.C. Downer, R.L. Fork, and C.V. Shank, "Amplified Femtosecond Optical Pulses and Continuum Generation at 5 kHz Repetition Rate," *Opt. Lett.* 9:552-552 (1988).

and limits carrier transport.[33] We have used our recently constructed a tunable femtosecond laser system to perform direct measurements of the intervalley scattering times in AlGaAs.

Our cavity dumped femtosecond laser system produces pulses on the order of 75 fs in the wavelength range 580 to 670 nm using the dyes rhodamine 6G and DCM. This laser system has enabled us to directly measure the $\Gamma-L$ and $\Gamma-X$ scattering times using pump-probe absorption saturation techniques. Dramatic changes in the initial femtosecond scattering times were observed as the laser was tuned above and below the satellite valley minima. The dynamics were also investigated using differential transmission measurements using an optical multi-channel analyzer. Complimentary investigations were performed using a high repetition rate, copper-vapor-laser amplifier, pump-continuum-probe technique.[34] Future measurements will be performed on thinner, anti-reflection coated samples.

We have recently begun a collaboration with Professors Karl Hess at the University of Illinois and Christopher J. Stanton at the University of Florida. These investigations will combine experimental studies of carrier dynamics with Ensemble Monte Carlo theoretical techniques. Monte Carlo techniques permit a modeling of electron and hole distribution function dynamics where the effects of varying fundamental scattering rates and processes may be evaluated directly. Connection of experimental measurements with Monte Carlo should permit both a measurement of the time constants of fundamental scattering processes as well as the construction of a more comprehensive model for excited carrier dynamics in semiconductors. These studies are of direct relevance for high speed electronic and optoelectronic devices. Preliminary calculations show promising results and have been submitted to the Sixth International Conference on Hot Carriers in Semiconductors.[35]

1.10.2 Nonlinear Susceptibility

Femtosecond investigations of bandedge nonlinear dynamics in AlGaAs guided wave devices are directly relevant to the development of high-speed all optical switches. While previous studies of $X^{(3)}$ have been performed using four-wave mixing and interferometry, measurement of electronic nonlinearities were often complicated by the presence of parasitic thermal index changes.[36] We have recently developed a novel technique for nonlinear index measurements which employs time division interferometry and eliminates thermal parasitics. The system can detect phase shifts as small as $\lambda/1000$ and requires no active stabilization.[37]

Our synchronously-pumped, tunable femtosecond laser system was used for these investigations. Using the dye styryl 9, we obtained 430 fs pulses in the wavelength range 780 to 880 nm. A single mode ridge waveguide was designed with an absorption edge appropriate for these wavelengths. The

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33 M.A. Littlejohn, J.R. Hauser, T.H. Glisson, D.K. Ferry, and J.W. Harrison, "Alloy Scattering and High Field Transport in Ternary and Quaternary III-V Semiconductors," *Solid State Electron.* 21:107-114 (1978).

34 W.Z. Lin, M.J. LaGasse, R.W. Schoenlein, B. Zysset, and J.G. Fujimoto, "Femtosecond Studies of Excited Carrier Energy Relaxation and Intervalley Scattering in GaAs and AlGaAs," invited paper presented at the SPIE Symposium on Advances in Semiconductors and Superconductors, Newport Beach, California, March 1988.

35 C.J. Stanton, D.W. Bailey, K. Hess, M.J. LaGasse, R.W. Schoenlein, and J.G. Fujimoto, "Femtosecond Studies of Intervalley Scattering in GaAs and AlGaAs," submitted to the Sixth International Conference on Hot Carriers in Semiconductors.

36 G.I. Stegman, E.M. Wright, N. Finlayson, R. Zanoni, and C.T. Seaton, "Third Order Nonlinear Integrated Optics," *J. Lightwave Tech.* 6:953-970 (1988).

37 M.J. LaGasse, D.L. Wong, J.G. Fujimoto, and H.A. Haus, "Femtosecond pump probe interferometry," Proceedings of IQEC '88, Tokyo, Japan, July 1988, paper WG4, p. 538; M.J. LaGasse, K.K. Anderson, H.A. Haus, and J.G. Fujimoto, "Femtosecond All-Optical Switching in AlGaAs Waveguides Using a Single Arm Interferometer," *Opt. Lett.* 14 (6):314 (1989).

instantaneous nonlinear index as a function of wavelength detuning was measured. The results show a dramatic increase in the nonlinear susceptibility as the laser is tuned towards the bandedge. However, the linear absorption is also increasing rapidly in this wavelength regime.

For larger intensities, two photon absorption becomes dominant. Also, self phase modulation increases the spectrum of the laser pulse and dispersion becomes a limiting factor. Our measurements indicate that AlGaAs waveguides may not satisfy the requirements for a practical ultrafast all-optical switching material. In particular, the nonlinearity is not large enough, and scaling of intensity and length is complicated by two-photon absorption.

Future measurements will apply our techniques to characterizing quantum well waveguides where quantum confinement effects may enhance the ultrafast nonlinear index. Systematic studies using time domain techniques can be applied to measure nonlinear index, two photon absorption as well as loss and dispersion. A detailed knowledge of these parameters is essential for predicting waveguide switching behavior and modeling.

1.11 Femtosecond Electron Dynamics in Metals

Sponsors

AT&T Bell Foundation
National Science Foundation
(Grant ECS 85-52701)
Joint Services Electronics Program
(Contracts DAAL03-86-K-0002
and DAAL03-89-C-0001)
U.S. Air Force - Office of Scientific Research
(Contract F49620-88-C-0089)

Project Staff

Robert W. Schoenlein and Professor James G. Fujimoto in collaboration with Dr. Gary Eesley and Dr. Wes Capehart

Femtosecond optical studies of electron dynamics in metals provide information about fundamental physical processes in high density systems. Since the principal constituents of devices are metals and semiconductors, an understanding of electron dynamics in metals and at metal-semiconductor interfaces is relevant to the development of high speed electronic and optical devices.

In contrast to semiconductors, relatively few investigations have been performed in metals. Because of the high electron densities in these materials, nonequilibrium processes and excited state dynamics occur on a very short time scale. In addition, changes in optical properties associated with these dynamics are extremely small. Thus, transient spectroscopy in metals has required the development of new femtosecond laser technology and measurement techniques which provide high temporal resolution and sensitivity.

Nonequilibrium electron heating in metals has been investigated using time resolved differential reflectivity measurements.[38] In these studies, a femtosecond laser pulse heats the electron distribution to a temperature in excess of the lattice temperature. This occurs because the heat capacity of the electron gas is much less than that of the lattice. Energy relaxation of the electron distribution occurs through electron-phonon interaction. This is investigated in noble metals by monitoring optical transitions from the *d*-bands to electronic states near the Fermi level. We have demonstrated nonequilibrium electron heating using time resolved pump and continuum probe reflectivity lineshape measurements on a femtosecond time scale. Electron-phonon energy transfer times of 1-2 ps were observed.

In addition to reflectivity studies, we are currently working in collaboration with researchers from General Motors Research Laboratories to investigate the transient dynamics of image potential states in

[38] R.W. Schoenlein, W.Z. Lin, J.G. Fujimoto, and G.L. Eesley, "Femtosecond Studies of Nonequilibrium Electronic Processes in Metals," *Phys. Rev. Lett.* 58:1680 (1987).

metals.[39] Image potential states are formed by the Coulombic attraction between an electron at the surface of a metal, and its image charge inside the solid. The electronic wavefunction is localized outside the metal and the states occupy a Rydberg-like series of energy levels approaching the vacuum potential. These states are of particular interest because they represent two-dimensional electron confinement, analogous to quantum-well structures in semiconductors, but are not subject to phonon scattering and other processes associated with the pulse material.

In order to study the dynamics of these states, we are developing new measurement techniques which combine femtosecond pump probe with photoemission spectroscopy. This enables us to investigate transient electron dynamics in high density materials. Lifetime measurements of the $n = 1$ image potential state in Ag(100) have been performed using these techniques.[40]

Ultraviolet ($\sim$ 4 eV) femtosecond pulses, generated via frequency doubling, are used to populate the image potential states. Electrons are then photoemitted from the states by a second visible ($\sim$2 eV) femtosecond pulse delayed in time. The photoemitted electrons are collected and the energy spectrum is measured using a cylindrical mirror analyzer. This approach provides background-free time-resolved measurements of the image-potential state energy distribution. The $n = 1$ state in Ag(100) is observed to have a lifetime of $\sim$ 30 fs. Development of a broadband femtosecond pulses in the UV using frequency doubling and continuum generation will enable us to extend these studies to other material systems.

1.12 Laser Medicine

Sponsors

National Institutes of Health
(Grant 5-RO1-GM35459)
Massachusetts General Hospital
(Office of Naval Research
Contract N00014-86-K-0117)

Project Staff

Beat Zysset, Jhypyng Wang, Robert W. Schoenlein, Professor James G. Fujimoto in collaboration with Professor Thomas Deutsch, Professor Reginald Birngruber, and Professor Carmen Puliafito.

1.12.1 Time Resolved Studies and Biological Effects of Ultrashort Pulsed Breakdown

Laser induced optical breakdown, or photodisruption, using ultrashort laser pulses has recently achieved widespread clinical use for ophthalmic surgery in the anterior eye.[41] The use of picosecond pulse durations for laser surgery can result in a significant enhancement of the desired incision effects while reducing unwanted collateral tissue damage. In contrast to nanosecond pulses, the high peak intensities necessary for the initiation of laser induced optical breakdown may be achieved with reduced pulse energies.

We have performed studies of ultrashort pulse breakdown using single 40 ps Nd:YAG laser pulses. Time resolved measurement techniques were developed and applied to perform a comprehensive study of the temporal and spatial dynamics of the plasma

39 D. Straub and F.J. Himpsel, "Identification of Image-Potential Surface States on Metals," *Phys. Rev. Lett.* 52:1922-1924 (1984).

40 R.W. Schoenlein, J.G. Fujimoto, G.L. Eesley, and T.W. Capehart, "Femtosecond Studies of Image-Potential Dynamics in Metals," *Phys. Rev. Lett.* 61:2596 (1988).

41 Fankhauser, P. Roussel, J. Steffen, E. Van der Zypen, and A. Cherenkova, "Clinical Studies on the Efficiency of High Power Laser Radiation Upon Some Structures of the Anterior Segment of the Eye," *Int. Ophthalmol.* 3:129-139 (1981); J.J. Aron-Rosa, Aron, J. Griesemmann, and R. Thyzel, "Use of the Neodymium YAG Laser to Open the Posterior Capsule After Lens Implant Surgery: A Preliminary Report," *J. Am. Intraocul. Implant Soc.* 6:352-354 (1980).

formation, shock wave, and cavitation processes which accompany the breakdown process.[42] Tissue effects were investigated using the cornea endothelium in vitro.[43] Comparison with physical measurements suggests that endothelial cell damage is mediated by shock wave and cavitation processes while incisions confined to focal region of the laser beam are produced by the laser induced plasma. Systematic studies were performed to determine the energy scaling behavior of the tissue effects and to quantify the minimum "safe" distance for clinical photodisruption near sensitive stuctures. A minimum damage range of less than 100 μm was achieved for pulse energies of 8 μJ.

In contrast to nanosecond pulses or multiple pulse mode-locked picosecond pulse trains, the use of single picosecond pulses permits a significant reduction in breakdown energy threshold and collateral damage. These results suggest the possibility of developing surgical techniques for producing highly localized incisions of intraocular structures.

1.12.2 Corneal Ablation with Ultrashort Optical Pulses

Pulsed laser light can ablate the cornea at wavelengths at which the cornea is nominally transparent if the intensity at the corneal surface is sufficiently high that optical breakdown occurs.[44] We have investigated corneal ablation at visible wavelengths at pulse durations of 65 fs, 100 fs, 1 ps, 30 ps, and 8 ns. Spot size (diameter) at the corneal surface was approximately 25 μm. The threshold energy for ablation was proportional to the square root of the pulse duration and varied from 2.5 μJ at 100 fs to 500 μJ at 8 ns. Etch depth per pulse at the threshold energy was less than 0.1 μm with picosecond and femtosecond pulses, compared to 1 μm with 8 ns pulses. At 8 ns the excision walls were very ragged or irregular, disorganization or denaturation of collagen fibrils extended 10 μm from the excision edges at all pulse energies. With picosecond and femtosecond pulses the excision walls were relatively straight and smooth. At energies near the ablation threshold, denaturation of collagen fibrils did not extend more than 1 μm from the excision edges and disorganization of collagen fibrils was slight. Picosecond and femtosecond lasers at low pulse energies produce excisions of sufficiently high quality that they can be regarded as possible alternatives to excimer lasers for corneal surgery. We speculate that ultrashort-pulsed lasers might, in addition, have advantages over conventional ophthalmic Nd:YAG lasers for selected intraocular applications.

1.12.3 Femtosecond Retinal Injury

Initial studies of femtosecond retinal injury showed that minimal retinal effects can be produced only with peak intensities higher than 10^{11}W/cm^2 and that, in contrast to laser injuries produced by longer pulses, exposures of more than 100 times threshold did not produce significantly more severe lesions or hemorrhage.[45] This unique finding suggests that femtosecond pulses may be useful in producing highly localized laser effects in the posterior segment of the eye; we conducted experiments to characterize the biophysical basis of this effect. Initial investigations showed that laser pulses with intensities higher than 10^{12}W/cm^2 cannot be transmitted through the transparent media of the eye because of the presence of nonlinear optical

[42] B. Zysset, J.G. Fujimoto, and T.F. Deutsch, "Time-Resolved Measurements of Picosecond Optical Breakdown," *Appl. Phys. B* 48:139-147 (1989).

[43] B. Zysset, J.G. Fujimoto, C.A. Puliafito, R. Birngruber, and T.F. Deutsch, "Picosecond Optical Breakdown: Tissue Effects and Reduction of Collateral Damage," *Lasers in Surgery and Medicine,* in press.

[44] D. Stern, R.W. Schoenlein, C.A. Puliafito, E.T. Dobi, R. Birngruber, and J.G. Fujimoto, "Corneal Ablation by Nanosecond Picosecond, and Femtosecond Lasers at 532 and 625 nm," *Archives of Ophthalmology,* in press.

[45] R. Birngruber, C.A. Pulifito, A. Gawande, W.-Z. Lin, R.T. Schoenlein, and J.G. Fujimoto, "Femtosecond Laser-Tissue Interactions: Retinal Injury Studies," *IEEE J. Quantum Electron.* QE-23:1836-1844 (1987).

effects such as self focusing or continuum generation at such high intensities. Since nonlinear damage limiting effects do not occur with propagation of laser pulses in gases, we performed lensectomy, vitrectomy, and total air-fluid exchange in 18 rabbit eyes. In some eyes, fibrin membranes were created adjacent to the retinal surface, and the ability of femtosecond pulses to cut these membranes was studied. A total of 222 laser exposures were made using pulse energies between 0.6 μJ and 125 μJ using laser pulses of 100 fs and 1 ps in duration. Membranes could be cut with pulse energies of 1 to 10 μm; morphologic investigations revealed minmal damage to adjacent neural retinal structures. We conclude that femtosecond pulses can be coupled to nominally non-absorbing tissue by nonlinear absorption mechanisms and are potentially useful for tissue cutting with high spatial confinement.

1.12.4 Femtosecond Optical Ranging of Corneal Incision Depth

Excimer laser ablation has been proposed as a technique for keratorefractive surgery. Clinical acceptance of linear-incision laser keratectomy may depend on the availability of a method for accurately and noninvasively monitoring incision depth during the ablation process. We have developed a femtosecond optical ranging techqnique for measurement of corneal incision depth.[46] This technique uses nonlinear optical cross-correlation to determine the time-of-flight of an ultrashort laser pulse between the anterior corneal surface and the bottom of the keratectomy incision. Longitudinal and transverse resolution are estimated to be 5 μm and 10 μm, respectively.

1.13 A New Conjugate Gradient-Type Algorithm for Rate Equations

Sponsor

Lawrence Livermore National Laboratory
(Subcontract BO48704)

Project Staff

Sumanth Kaushik, Professor Peter L. Hagelstein

Since the amplifier for the proposed x-ray laser is a laser induced plasma, it is imporant to properly characterize the behavior of this plasma in order to have reliable estimates of gain. One important parameter in computing gain is the relative population of the various atomic levels of the amplifying media.

The computation of the relative population of the atomic states in a nonequilibrium plasma requires the solution of a large set of population rate equations.[47] Since analytical solutions are impossible and in most cases, numerical computation is often the only alternative. A numerical solution to these rate equations requires the inversion of large matrices that arise from the numerical discretization of rate equation. Since computational resources in general are usually limited, an efficient method to invert these large matrices is sought.

Hitherto, conjugate gradient methods[48] have not been successfully applied to these rate matrices. The rate matrices are typically very dense and stiff; hence, modern sparse matrix techniques were not particularly successful. However, as a result of our research efforts, we have managed to develop a conjugate gradient-type algorithm which can efficiently solve these large stiff matrix systems. Our algorithm uses existing sparse matrix

46 D. Stern, W.-Z. Lin, C.A. Puliafito, and J.G. Fujimoto, "Femtosecond Optical Ranging of Corneal Incision Depth," *Inv. Ophthal. Vis. Sci.* 30:99-104 (1989).

47 D. Mihalas and B. Mihalas. In *Foundations of Radiation Hydrodynamics,* 309-410. Oxford University Press, 1984.

48 D. Kershaw, *J. Comput. Phys.* 26:43-65 (1978).

methods with certain important and original modifications.[49]

We report the success of the preconditioned biconjugate gradient (PBCG) algorithm in solving the matrix equations resulting from the discretization of systems of population rate equations which arise in nonequilibrium kinetics modeling. The success of the PBCG can be attributed to two main ideas. First, the singularity of the rate matrix resulting from population conservation requirement was removed through a reduction of matrix order so as to improve the condition number of the matrix. Second, an efficient preconditioner was found to reduce the eigenvalue spread of the rate matrix. The preconditioning matrix was selected on the basis of retaining the largest few rates in each column of the well conditioned rate matrix. This preconditioner, along with the reduced rate matrix, enabled the PBCG to converge very rapidly as to make it an attractive alternative to standard direct methods.

The preconditioned BCG algorithm is given by:[50]

$$\alpha'_k = \frac{[(U^T)^{-1} \cdot s_k]^T \cdot [L^{-1} \cdot r_k]}{q_k^T \cdot A \cdot p_k}$$

$$x_{k+1} = x_k + \alpha'_k \cdot p_k$$

$$r_{k+1} = r_k - \alpha'_k A \cdot p_k$$

$$s_{k+1} = s_k - \alpha'_k A^T \cdot q_k$$

$$\beta'_k = -\frac{[(U^T)^{-1} \cdot s_{k+1}]^T \cdot [L^{-1} \cdot r_{k+1}]}{[(U^T)^{-1} \cdot s_k]^T \cdot [L^{-1} \cdot r_k]}$$

$$p_{k+1} = U^{-1} \cdot L^{-1} \cdot r_{k+1} - \beta'_k p_k$$

$$q_{k+1} = (L^T)^{-1} \cdot (U^T)^{-1} \cdot s_{k+1} - \beta'_k q_k$$

with the initial conditions $r_o = s_o = p_o = q_o = b - A \cdot x_o$, and where x_o is the initial guess vector. This algorithm implements the biconjugate gradient method on the system

$$L^{-1} \cdot A \cdot U^{-1} \cdot (U \cdot x) = L^{-1} \cdot b$$

which is equivalent to $\bar{\bar{A}} \cdot \bar{x} = \bar{b}$.

This algorithm was tried on our test matrix. As seen in figure 3, the results were quite successful. There are two curves shown in figure 3. One curve shows the iteration plot for a poor initial guess ($\|x_o - x^*\|_2 = 7.0$); the other curve shows the iteration plot for a good initial guess $\|x_o - x^*\|_2 = 9.6 \times 10^{-1}$. In both cases, $\|x^*\|_2 = 1$. The algorithm converged in less than thirty iterations in the first case and in as little as eight iterations for the second case. In transient NLTE calculations, the initial guess at timestep n+1 is the solution at timestep n, and this guess tends to be closer to the final solution at timestep n+1, on average, than our "good initial guess" example here.

The success of the PBCG over Gaussian elimination must be compared in terms of both memory used and operation count. In order to efficiently use memory, the non-zero matrix elements of the matrices A, L and U were all stored as lists and the coordinates corresponding the non-zero matrix elements were stored in a integer array. The memory required by the PBCG algorithm as implemented requires:

$$M_{PBCG} = fN^2 + 2mN + \frac{fN^2}{4} + \frac{mN}{2} + 11N$$

where fN^2 is for A stored as a list; where 2mN is for the L and the U stored as a list; 11N is for the vectors and the remaining for the integer index vectors need for the list. As before, f denotes the fill factor. For the test

[49] S. Kaushik and P. Hagelstein, "The Application of the Preconditioned Biconjugate Gradient Algorithm to NLTE Rate Matrix Equations," submitted to *J. Comput. Phys.* (1988).

[50] Z. Mikic and E.C. Morse, *J. Comput. Phys.* 61:154-185 (1985).

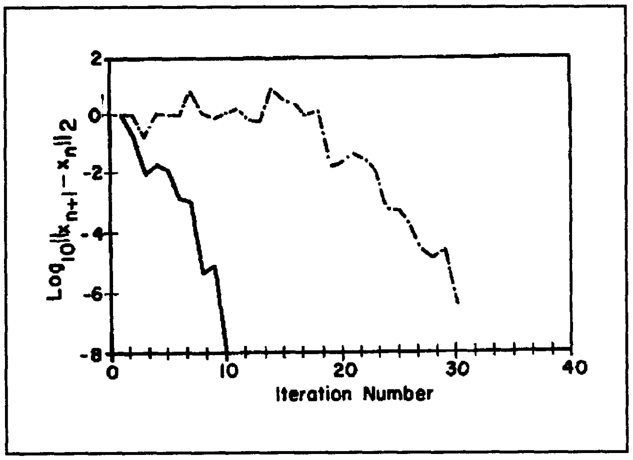


Figure 3.

matrix above ($f = 19$ percent, m=7, N=258), the savings in memory is 70 percent.

To compare speed, the number of floating point multiplications and additions required for Gaussian elimination is roughly

$$N_{Gauss} \approx f\frac{N^3}{3} + \frac{N^2}{2}$$

under the assumption that the coding is such that all "zero" multiplies and additions are avoided. This optimistic limit is not usually obtained in hand-coded Gaussian elimination routines. However, it could in principle be obtained on some future computer whose architecture permitted fast concurrent zero checks. The PBCG iterations require roughly

$$N_{PBCG} \approx k(2fN^2 + 2mN + 8N)$$

additions and multiplications under similar assumptions. There is some time required for setup, but we have found this to be rather small in comparison with the time spent on the iterations. The ratio of operations for the two methods is approximately

$$\frac{N_{PBCG}}{N_{Gauss}} \sim \frac{6k}{N}$$

for a matrix with a moderately large fill factor. In this case, the full matrix multiplies of the PBCG method dominates the computation time. For the examples considered above, the ratios are 0.7 and 0.2 for the poor and good initial guesses, respectively. This estimate is a rather ideal operation count estimate, and in our experience so far, the observed ratio of run times tends to favor the PBCG by larger factors.

1.14 Large Nd:Glass Laser System and X-Ray Laser

Project Staff

Martin H. Muendel, Professor Peter L. Hagelstein

We are designing a small Nd:glass laser system to use as a driver for a small x-ray laser.[51] The system will consist of an mode-locked, Q-switched Nd:YLF oscillator and two-pass Nd:glass preamplifier supplying pulses of 100 mJ, and two Nd:glass slab power amplifiers in parallel, each generating 5 J output. The oscillator and preamplifier have recently been installed, and the slab amplifiers will be built over the Summer of 1989 in collaboration with the Lawrence Livermore National Laboratory.

In general, the slab amplifier design follows designs developed at Livermore and at Stanford over the past few years. Each slab measures about 30 cm in length by 20 cm width by 1 cm thickness. The laser beam goes through the slab lengthwise, following a zig-zag path in which it reflects internally on the slab faces. This zig-zag path evens out any gain nonuniformities and thermal distortions across the slab thickness. Each slab is pumped by four large xenon flashlamps in a silvered pumping chamber; the energy stored in each slab is 0.25 J per cubic cm, giving a gain coefficient of 0.05 per cm. The beam passes through the slab four times, giving a net gain of 100.

Because of the small size of the system and the fast cooling time of the slabs, the system will be able to fire once every ten seconds, a

[51] P.L. Hagelstein, "Short Wavelength Lasers: Something New, Something Old," Proceedings of the *Conference on Short Wavelength Coherent Radiation,* Cape Cod, MA (1988).

very high repetition rate by the standards of most x-ray lasers drivers. Because thermal distortion in the slabs will be so small, the output beams will be of high quality and the danger of optical damage to the slabs will be minimal.

Several aspects of the slab design are novel. The output will be in short trains of 100-picosecond pulses, the shortest pulses that anyone yet has tried to amplify in a slab. The high peak intensities in the short pulses create severe problems with nonlinear effects which we will have to counteract with spatial filters and some other optics. Amplifying the whole train of pulses will cause some problems. Also, using the slab as a power amplifier rather than as an oscillator or a regenerative amplifier is unusual and will require us to use several separate beam passes through the slab. Finally, a system to cool the slab must also be designed: we are planning to use air cooling rather than water, the usual choice. This choice will avoid many problems such as the gradual degradation of a phosphate glass surface in contact with water.

The output of the driver laser will be focused to a line 1 cm long by about 50 microns wide on a target of molybdenum or similar metal. The molybdenum will be ionized by the driver pulse to form a plasma of nickel-like ions. Electron-ion collisions will then cause excitation of the nickel-like ions, leading to a population inversion and gain along the length of the line. The target will be situated in a reflective cavity (multilayer or whisper gallery mirrors), giving x-ray laser oscillator action. This scheme will generate an output beam at 194 Å; we are also considering schemes using plasmas of, for example, Nd-like uranium,[52] to reach wavelengths as short as 70 Å.

1.15 Whisper Gallery Mirrors

Project Staff

John Paul Braud, Professor Peter L. Hagelstein, Tsen-Yu Hung

Research continues on the subject of whisper gallery mirrors (WGMs) for use in the soft x-ray and extreme ultraviolet regimes. In the language of ray optics, a WGM is a concave structure which, by means of a series of relections at small angles of incidence, can deflect radiation through a large total angle. At the wavelengths of interest, many materials have a refractive index less than unity. This allows for the possibility of WGMs which function on the principle of "total internal reflection" from a dielectric surface.

The principle limitation on the overall reflectivity of such a WGM arises from photoabsorptive loss in the dielectric. Because the resulting mirror performance is so strongly dependent upon the choice of dielectric material, we have made extensive studies as to which materials should be most appropriate.[53] Experimentally measured optical constant data were collected from the literature and analytical checks were then made upon them using Kramers-Kronig relations and sum rules. These results were used to give estimates of WGM performance. At the particular wavelengths of interest to us for applications to x-ray lasers, i.e., around 200 Å, the resulting WGM reflectivities are comparable with those predicted for normal-incidence multilayer mirrors.

Interestingly, the two materials yielding the highest potential reflectivities between 200 Å and 300 Å were argon and krypton. This fact raised the question of whether it might be possible to construct a WGM, the surface of which is a layer of frozen inert gas. As it turns out, argon and krypton apparently can

[52] P.L. Hagelstein and S. Dalhed, "Strong Monopole Collisional Excitation in Highly Stripped Atoms," *Phys. Rev. A* 37:2227 (1988).

[53] A.V. Vinogrador, V.F. Koralev, I.V. Kozhevnikov, and V.V. Ristovalor, "Concave Rotating X-Ray Mirror: I," *Sov. Phys. Techn. Phys.* 30 (2):145 (1985); A.V. Vinogrador, V.F. Koralev, I.V. Kozhevnikov, and V.V. Ristovalor,"Diffraction Theory for Crazing Modes in Concave Mirrors and Resonators at X-Ray Wavelengths: II," *Sov. Phys. Tech. 30 (3):335 (1985).*

form very smooth layers when frozen. We envisage a mirror system in which this sort of frozen surface is surrounded by a sparse atmosphere of the same inert gas, with the system held to the temperature at which the surface and atmosphere are in equilibrium: sublimation from the surface would be balanced by adsorption (see figure 4). Although complicated, this sort of mirror might have a couple of practical advantages over its more conventional counterparts. First, this cryogenic mirror surface would be self-healing; any damage to it would be smoothed over by the processes of sublimation and adsorption. Second, so long as the inert gas atomosphere were kept fairly free of oxygen, this type of mirror would be immune to the formation of an oxide layer, a problem which often plagues optics in the extreme ultraviolet and soft x-ray regimes as a result of oxygen's high photoabsorption cross-section and its affinity for metal surfaces.

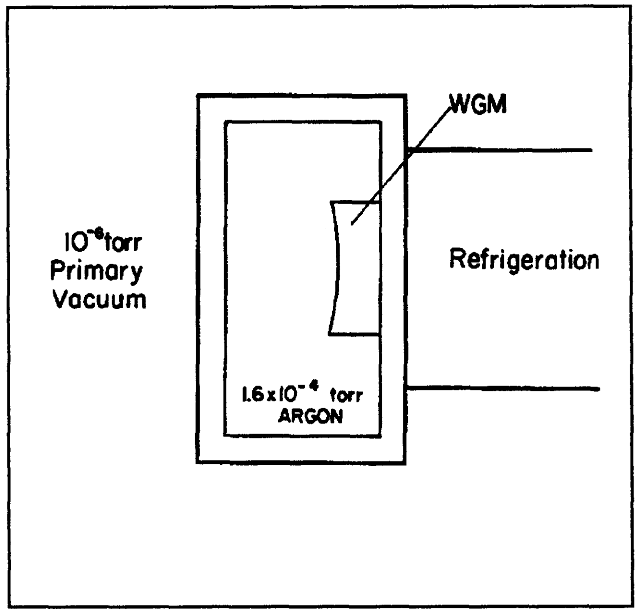


Figure 4.

1.16 X-Ray Detection Using Quantum Well Exciton Nonlinearity

Project Staff

Cris C. Eugster, Professor Peter L. Hagelstein

Research efforts continue on the development of a multiple quantum well x-ray detector. The detection method is based on the strong carrier density dependence of the "complex" index of refraction of the exciton absorption lines in GaAs/AlGaAs MQW structures.[54] Two related schemes have been developed in parallel which are based on 1) the scattering and 2) the reflection of a pulsed probe beam by the carrier distribution resulting from the absorption of a single energetic x-ray photon.

Extensive simulations have been conducted to characterize the various parameters of the devices. The results of these simulations have shown very high degrees of temporal resolution (< 50 psec) and spatial resolution (< 1 micron) for both schemes. The simulations have also verified the strong signal enhancement which occurs when the probe beam is tuned in the avalanche regime (at the knee of the heavy hole exciton line). In this regime, the absorption coefficient of the material increases with increasing carrier density thereby allowing the probe beam to preferentially create signal carriers in the region where the x-ray was absorbed.

At this point, plans are underway to fabricate and experimentally test some of these devices. Possible applications of such as fast high resolution x-ray detector include inertial confinement fusion studies, research in atomic physics of high density plasmas, in x-ray laser experiments, and as imaging instruments for microscopic and holographic applications.

[54] D. Chemla, D. Miller, A. Miller, P. Smith, A. Gossard, and W. Wiegman, "Room Temperature Excitonic Nonlinear Absorption and Refraction in GaAs/AlGaAs Multiple Quantum Well Structures," *IEEE J. Quantum Electron.* 20:265-275 (1984).

1.17 Study and Development of a Variational Method for Relativistic Atomic Structure Calculations

Project Staff

Leon M. Balents, Professor Peter L. Hagelstein

In non-relativistic quantum mechanics, calculation of energy levels of multi-electron atoms is done primarily in two basic ways. The differential equations involved (Hartree-Fock) can be solved numerically, or the variational principle can be employed to obtain better approximations to the exact eigenvalue. The latter method has the advantages both of speed and the possibility for greater insight. In the relativistic case, the variational method cannot be directly applied, since the Dirac hamiltonian is not bounded from below. Direct use of the Dirac hamiltonian results in the Brown-Ravenhall disease of variational collapse.[55] Quantum field theory yields the result that this problem can be overcome by the use of projection (Λ_{++}) operators that project out the negative energy states,[56] but the form of these operators is not well understood. Because of these problems, relativistic calculations of energy levels in multi-electron atoms are ordinarily done using numerical Dirac-Hartree-Fock (DHF) programs.[57] It now appears that a practical method of applying the variational method to relativistic problems has been found.[58] Through the use of a specific form of finite basic set, the DHF equations can be made to yield approximate eigenvalues, which, when minimized, appear to yield a variational principle for the Dirac hamiltonian.

If this method proves to be truly variational for all possibe forms of the wavefunction, it will provide a powerful means of performing atomic calculation. By introducing multiconfigurational states, the bounds on the actual energy levels will be computable to machine accuracy (up to uncertainties in Q.E.D. terms). If the essential portions of this procedure can be determined, it will lead to a greater understanding of the nature of the Dirac equation, with applications to many areas.

Our work aims to investigate the validity of extensions of this scheme to multi-configurational calculation - something not yet attempted in the literature. A code has been constructed to perform calculations in simple cases of both single and multi-configurational relativistic HF, and preliminary results are promising. These results help to develop understanding of the method's mechanism, and may have an application in computing later perturbative QED corrections by defining analytic projection operators.[59]

1.18 Large Coherency and Cold Fusion

Project Staff

Professor Peter L. Hagelstein

Some recent and controversial experiments suggest that fusion may occur near room temperature. By itself, such a result is not revolutionary in that it is known that cold muons can catalyze fusion at room temperature.[60] What is remarkable is that reactor level fusion heat output has been claimed by Fleischmann and Pons. To date, this claim

[55] G.E. Brown and D.E. Ravenhall, *Proc. Royal Soc. London,* Ser. A. 208:552 (1951).

[56] J. Sucher, "Foundations of a Relativistic Theory of Atomic Structure," In *NATO Advanced Study Institute on Relativistic Effects in Atoms, Molecules, and Solids.* New York: Plenum Press, 1981.

[57] I.P. Grant, *Proc. Royal Soc. London,* Ser. A. 262:555 (1961).

[58] S.P. Goldman and A. Dalgarno, *Phys. Rev. Lett.* 57 (4):408 (1986).

[59] L. Balents, S.B. thesis, Dept. of Physics, MIT, 1989.

[60] M. Fleischmann and S. Pons, *J. Electroanalytical Chem.* 261:301 (1989).

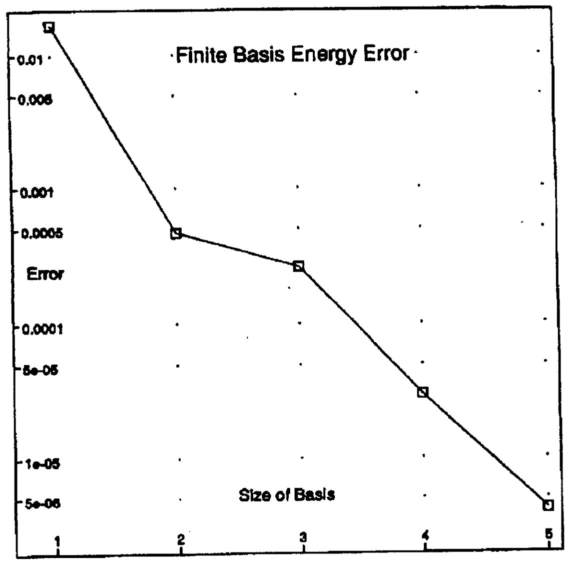


Figure 5.

has been received with much skepticism by the scientific community due to the lack of any known mechanism by which this could occur.

We have mused how such a fantastic claim could possibly be reconciled with known laws of physics, under the assumption that healthy skepticism be provisionally suspended temporarily.

If the highly exotic (and possibly nonexistent) reaction D + D → ^{4}He + lattice energy could occur, and if it occurred in a lattice which was relatively unperturbed, then we propose that it might go in a coherent fashion. Conventional D + D reactions are adequately described using a binary picture and perturbation theory. Phonon emission from a binary reaction is essentially impossible, since it would be mediated by photon emission which is depressed by many orders of α. We picture this process as a collective process involving in excess of 10^9 particles, which emit 10^9 phonons for every fusion which occurs.

If we assume that a wavefunction describing this process were given by

$$\Psi = \sum_{n=0}^{N_{D/2}} c_n(t) e^{iEt/\hbar} \Psi_n$$

where Ψ_n is a macroscopic lattice wavefunction wherein n fusions occurred. The amplitudes obey

$$i\hbar \frac{\partial}{\partial t} c_n = \langle n \mid H_s \mid n-1 \rangle c_{n-1}$$

$$+ \langle n \mid H_s \mid n+1 \rangle c_{n+1}$$

One finds that if the matrix elements were constant, that the average rate of fusions is linear in matrix element

$$\gamma \approx 1.70 \frac{\mid \langle H \rangle \mid}{\hbar}$$

If the channels involving neutron and tritium emission are treated as incoherent, and if one makes the ansatz that the matrix elements for the incoherent processes are the same as for the coherent processes, then it follows that

$$\gamma_n \approx 1.38 \frac{\gamma^2}{\Gamma_n}$$

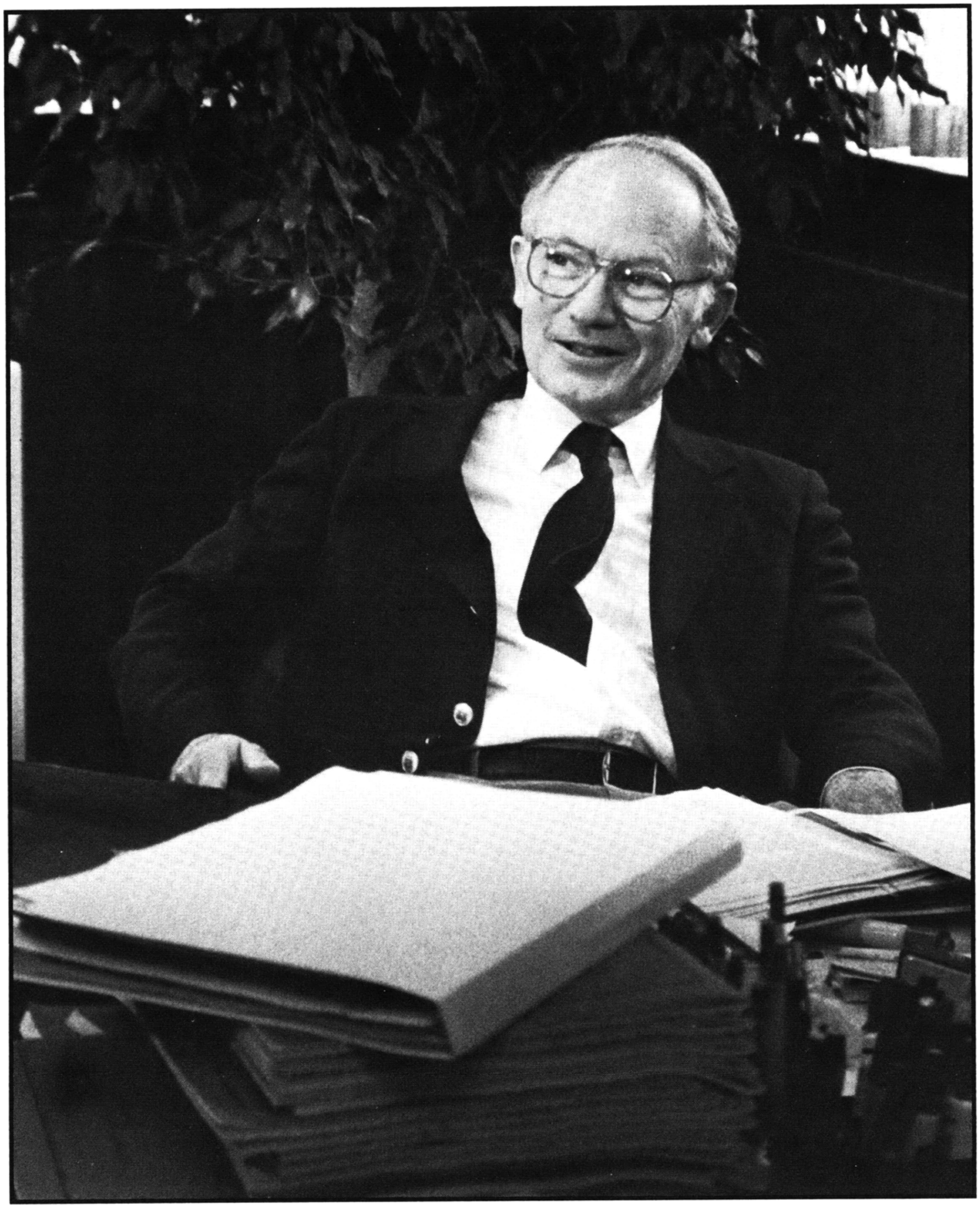
$$\gamma_T \approx 1.38 \frac{\gamma^2}{\Gamma_T}$$

where γ_n and γ_T are the neutron and tritium production rates, and Γ_n and Γ_T are dephasing rates. If we assume that dephasing occurs when the reactants are out of range of the strong force, we estimate that $\Gamma_n \approx 5 \times 10^{21}$ sec^{-1} and $\Gamma_T \approx 6 \times 10^{21}$ sec^{-1}. Fleischmann and Pons quote neutron and tritium emission numbers of 4×10^4 sec^{-1} and $1 - 2 \times 10^4$ sec^{-1}, respectively at 21.4 W/cm^3. The ansatz numbers from our simple coherent model are 1.4×10^4 sec^{-1} and 1.2×10^4 sec^{-1}, respectively.[61]

61 P.L. Hagelstein, "Neutron Tritium Production in Coherent DD Fusion," submitted to *Phys. Rev. Lett.* (1989).

Even though the scientific community is skeptical about the results of the new experiments, these results do seem to be consistent with a coherent picture of an exotic and possibly nonexistent reaction.

Professor Hermann A. Haus and graduate student Dilys L. Wong



Professor Peter A. Wolff